Phonon drag of electrons at high temperatures

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Abstract

Temperature-dependent thermoelectric power (TEP) in semiconductor crystals and crystal structures containing an electron-vibrational centers (EVC) was investigated in this article. The TEP contain narrow pius at Debye temperatures for different phonons. In thin epitaxial layers on substrates such pius exist at Debye temperatures of substrates phonons. These pius existence imposable to explain on basis of known TEP theory but it may be explained by the phonon drag of electrons (PDE) effect. In this connection there is the necessity to change traditional point of view on the PDE effect existence only at low temperatures and expand the PDE theory on case of strong electron-phonon coupling provided by EVC at high temperatures.

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I. INTRODUCTION

Thermoelectric power phenomenon was found out in 1821. This phenomenon arises in current-conducting materials at presence of temperature gradient. This physical phenomenon is caused by diffusion of mobile charges carriers in volume of a material under temperature gradient and is considered to be proportional to the temperature gradient. Factor of proportionality consist of two components which take into account the contributions of electrons (α_n) and holes (α_p) [1] :

$$\alpha = \alpha_n + \alpha_h. \tag{1}$$

If relaxation time of wave pulses for electrons and holes depend on energy (E) as E^r then in nondegenerated semiconductor

$$\alpha_n = -\frac{k}{|e|}(r + \frac{5}{2} - \frac{F_e}{kT}), \qquad \alpha_h = +\frac{k}{|e|}(r + \frac{5}{2} - \frac{F_h}{kT}), \tag{2}$$

where k - Boltzman constant, e - electron charge, r - dissipation parameter, F_e and F_h - Fermi levels for electrons and holes, T - temperature. In some semiconductor materials at low temperatures the experimental magnitude of TEP exceed the magnitude which was predicted on basis of the TEP diffusion theory. This disagreement between theory and experiment was explained by another physical phenomenon - phonon drag of electrons (PDE). The PDE represents the additional contribution in thermoelectric power. The PDE component is not connected with diffusion of mobile charges carriers but is caused by their moving under phonons flow action due to electron-phonon coupling.

For the first time L. Gurevich has predicted the PDE existence in metals and has specified the opportunity of the phenomenon existence in semiconductors [2]. G. Pikus investigated this phenomenon theoretically in semiconductors and in 1951 has deduced the formula for PDE which arise as a result of interaction of incoherent phonons flow with mobile charge carriers. He has shown that the magnitude of the PDE depend on the relation between relaxation times for wave pulses of phonons and electrons and has come to a conclusion about insignificant magnitude of the effect. However it was shown experimentally that the PDE in semiconductors can many times over exceed the magnitude which predicted by diffusion theory of thermoelectric power [3] - [5]. It was established that the PDE is caused by flow of those phonons which interact with electrons [3, 6, 7]. Or else, the PDE arises at presence of coupling between electrons and phonons.

The PDE component of TEP which take into account phonons flow interaction with mobile electrons may be described by coefficient

$$\alpha_{ph} = (\frac{k}{e})\frac{m^* v^2}{kT} \cdot \frac{< \tau_{ph} >}{< \tau >} \tag{3}$$

where $< \tau_{ph} >$ - middle time for phonons wave pulse relaxation, $< \tau >$ - middle time for electrons wave pulse relaxation, m^* - fictitious mass and v - speed of movable charge carriers [7]. In accordance to counting the long-wave acoustical phonons participation in the PDE effect the coefficient α_{ph} depend from temperature as $T^{-7/2}$ and decrease at heating.

The PDE effect was observed at temperatures above 70 K. Ones came to a conclusion that given effect is impossible to detect at T > 70 K. Absence of the given effect at T > 70 K was explained by weak electron-phonon coupling. Therefore the semi-conductor materials containing electron-vibrational centers (EVC) are attractive for supervision of the PDE effect [8] because inherent oscillations of such centers are capable to couple with electrons, phonons and hence to supply strong coupling between electrons and phonons.

The purpose of the given work is to research the phonon drag of electrons in crystals and crystal structures containing an electron-vibrational centers.

II. EXPERIMENTS, RESULTS AND DISCUSSION

A. Description of experiments

Industrial polished semiconductor plates by thickness 200 microns were used in our experiments. The semiconductor samples contained local electron-vibrational centers with representative strong electron-phonon coupling.

GaP samples with impurities of aluminum GaP(Al) or sulphur GaP(S) were investigated. Impurity concentrations of Al or S was equal to about $5 \cdot 10^{15}\text{cm}^{-3}$. Such impurities were chosen because atoms Al and S have masses vastly exceed mass of atom Ga. This promote emergence of electron-vibrational centers. Before measurements the samples were subjected to heating in vacuum within 5 minutes at temperature T = 600 K and cooled up to room temperature within 0.2 minutes to activate EVC.

Samples of Si, InSb, InAs were irradiated by fast electrons integer flow $\sim 10^{18}\ cm^{-2}$ with energy 1 MeV to introduce EVC. Silicon samples with impurities of phosphorus ($\approx\ 5 \cdot 10^{15}\text{cm}^{-3}$) and oxygen ($\approx\ 10^{18}\text{cm}^{-3}$) : Si(P,O) were used. Concentration of oxygen impurity

in Si was determined on basis of the data about distinctive for oxygen impurity optical absorption in spectral band near 9 mkm [9]. The main type of EVC in silicon samples was A-center which represent association of impurity oxygen atom with vacancy. In accordance to results of IR absorption spectra measurements the concentration of A-centers was equal to $\approx 10^{15} cm^{-3}$ and the Huang-Rhyce constant [10] for electron-phonon interaction $S \simeq 5$. Single crystals of InSb and InAs were investigated. Initial concentration of mobile electrons (before irradiation by fast electrons) at room temperature was equal $1.68 \cdot 10^{14} cm^{-3}$ for InSb and $6 \cdot 10^{12} cm^{-3}$ for InAs. Epitaxial layers of InSb and InAs with thickness up to 20 mkm on semiinsulator GaAs substrates with thickness 200 mkm were investigated else. Concentration of mobile electrons in initial InSb layer (before irradiation by fast electrons) was equal to $3 \cdot 10^{16} cm^{-3}$. Concentration of mobile electrons in initial InAs layer was equal to $3 \cdot 10^{16} cm^{-3}$, but after the irradiation it was near to inherent concentration. Supposedly EVC in the samples were formed by impurity oxygen atoms which usually are contained in significant concentrations in these semiconductor materials.

We investigated the carbon nanotube films with thickness $\simeq 0.1$ mkm with surface resistance $\cong 10^3\ Ohm \cdot cm^{-2}$ on quartz substrate. These samples were fabricated by dispersion of graphite single crystal by electron beam in vacuum [11]. Carbon nanotube films where consisted from single walled nanotubes. Diameter of nanotubes was about 10 nm. The nanotubes were oriented along normal to substrate surface and form two-dimensional regular structure. In accordance to investigated IR reflectance spectra and activation energies of resistivity the EVC in these samples were formed by carbon atoms. Supposedly the EVC existence is connected with variation of nanotube chirality.

The TEP were measured at temperatures is higher 77 K where the PDE existence was considered as impossible. Temperature difference between electrical contacts to every sample was not more than 3K, probable inaccuracy at temperature measurements was not more than 0.2 K and at voltage measurements it was not more than 10^{-6} V.

B. Experimental results

Typical temperature dependency of thermoelectric power for GaP(S) samples is shown in Fig. 1 by curve A. Curve A keeps pius which marked by arrows and by letters. Temperature dependency of the TEP for GaP(Al) is like the curve A. Curve B in Fig. 1 is the TEP

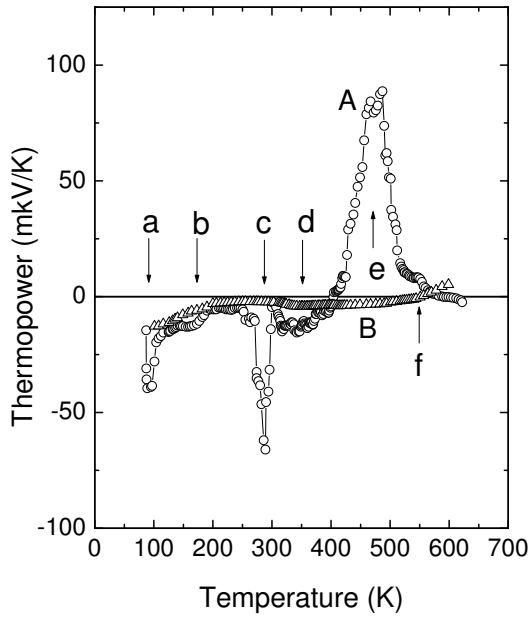

FIG. 1: Experimental temperature dependency of thermoelectric power in GaP with impurity of Sulphur (A) and calculated temperature dependency of thermoelectric power in GaP with close to inherent conductivity (B).

temperature dependency which was calculated by using Eqs. (1, 2) for GaP sample with close to inherent conductivity.

Typical temperature dependency of thermoelectric power for Si(P,O) samples is shown in Fig. 2 by curve A. This curve has pius specified by arrows and by letters. Curve B on Fig. 2 is temperature dependency of the TEP for Si single crystal with close to inherent conductivity which was calculated by using Eqs. (1, 2).

Temperature dependencies of the TEP for irradiated by fast electrons single crystals of InSb and InAs are shown in Fig. 3. Characteristic temperature dependency of thermoelectric power for irradiated epitaxial layer of InSb on semi-insulating GaAs substrate (InSb/GaAs) is represented in Fig. 4. Some pius on the curve are pointed out by arrows. Characteristic temperature dependencies of thermoelectric power for irradiated by fast electrons epitaxial layer of InAs on semi-insulating GaAs substrate (InAs/GaAs) are represented in Fig. 5.

Typical temperature dependencies of thermoelectric power for carbon nanotube film is shown in Fig. 6 by curve A. The TEP for silicon epitaxial layer by thickness 14 mkm on quartz substrate with impurity of Phosphorus ($\simeq 10^{14} cm^{-3}$) is shown in Fig. 6 by curve B

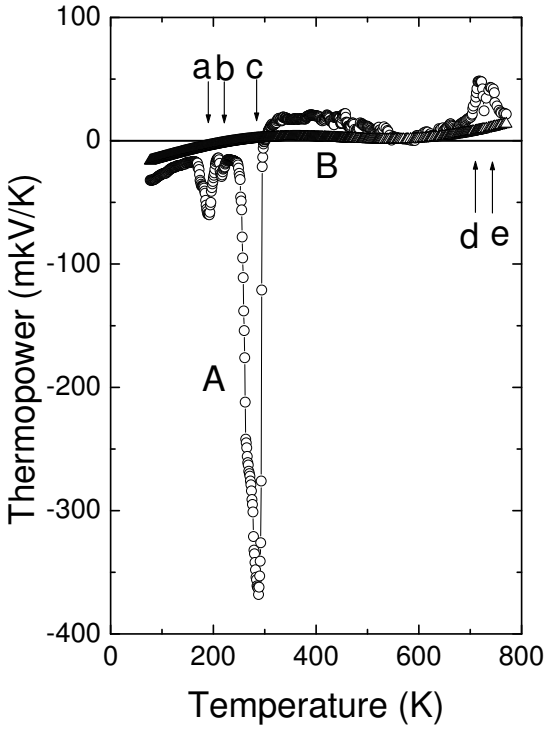

FIG. 2: Temperature dependency of thermoelectric power: experimental for Si containing A-centers (A) and calculated for Si with close to inherent conductivity (B).

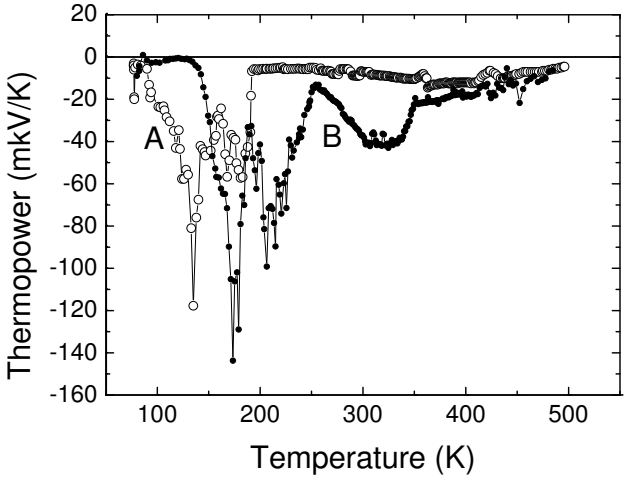

FIG. 3: Temperature dependency of thermoelectric power in irradiated by electrons single crystals InSb (A) and InAs (B).

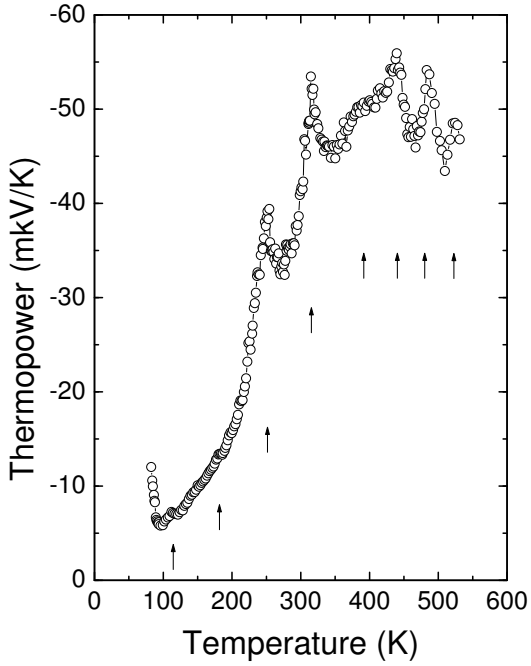

FIG. 4: Temperature dependencies of thermoelectric power in irradiated by fast electrons InSb epitaxial layer on semi-insulating GaAS substrate. Some pius of the dependencies are pointed out by arrows.

which is increased in 4 times in relation to valid curve.

C. Discussion

Curve A in Fig. 1 contain pius whose polarity agree with the diffusion TEP polarity. Pius a, b, c, f are situated at Debye temperatures of phonons in GaP [12]: 95 K (TA, 8.2 meV); 168 K (TA, 14.25 meV); 288 K (LA, 24.42 meV); 542 K (LO, 44.75 meV). Broad pius d ($\approx 345K$) and e ($\approx 475K$) may to be explained by combinations of phonons: (TA + TA, 28.6 meV) and (TA + LA, 38.67 meV). Curve B in Fig. 1 is the TEP which was calculated for GaP with close to inherent conductivity and without an account of interaction between electrons and phonons flow. This curve is smooth and does not contain any pius. Therefore located at Deby temperatures of phonons the narrow PDE pius in GaP(S) and GaP(Al) definitely may be connected with the EVC which are formed by impurity atoms of Sulfur or Aluminum.

Investigations of resistivity temperature dependencies, infra-red reflection spectra and the TEP temperature dependencies in GaP(Al) and GaP(S) show that impurity atoms of

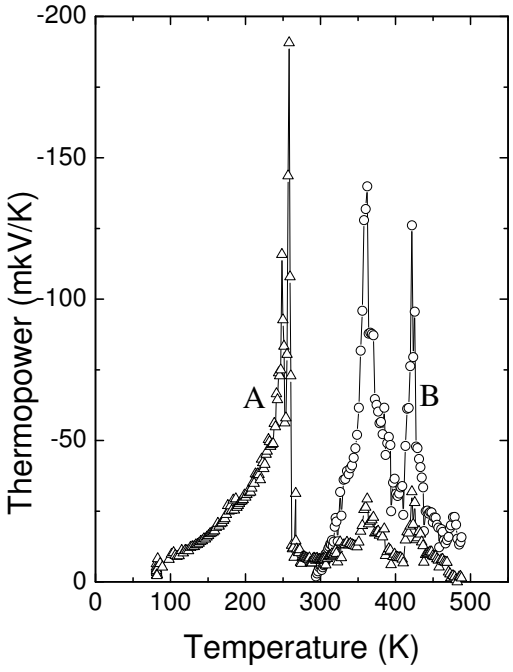

FIG. 5: Temperature dependencies of thermoelectric power in irradiated InAs epitaxial layer on semi-insulating GaAS substrate. Curve A was measured at heating but curve B was measured at cooling of the sample.

Al and S form the EVC in GaP due to which nuclei oscillations (inherent, I-oscillations) of Al and S actively interact with phonons and electrons and supply strong electron-phonon coupling. The I-oscillations can spread in crystals, interact with electrons and give rise to waves of I-oscillations and electrical currents. This is confirmed by particularities of the TEP. In connection with the given results it is possible to consider that I-oscillations of the EVC, which are formed by impurity atoms Al and S, are the reason of the narrow TEP peaks at Deby temperatures of phonons. It is possible to explain pius a, b, c, d by phonon drag of electrons and pius e, f by phonon drag of holes at Deby temperatures of the same phonons which effectively interact with EVC and supply strong electron-phonon coupling.

Curve A in Fig. 2 contain pius a, b, c which are situated at Debye temperatures of known characteristic acoustic phonons in Si whose wave vectors directed along certain directions in Brillouin zone [13]: <111>, 200.4 K (16.7 meV); <110>, 214.8 K (17.9 meV); <100>, 252 K (21.0 meV). Curve B on Fig. 2 is the TEP temperature dependence which was calculated for Si with close to inherent conductivity without any account of interaction between electrons

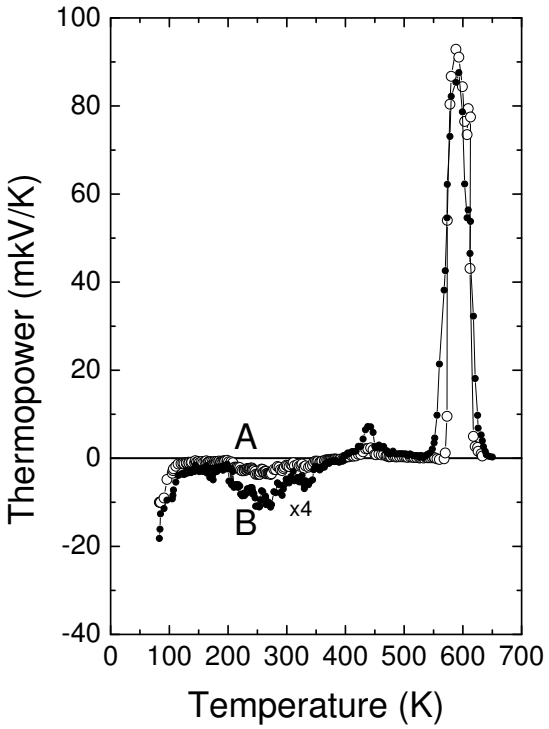

FIG. 6: Temperature dependency of thermoelectric power in carbon nanotube film on quartz substrate (A) and in silicon epitaxial layer on quartz substrate (B). Curve B is increased in 4 times in relation to valid curve.

and phonons flow. Comparison of curves A and B in Fig. 2 allows to connect the pius a, b, c in curve A with presence of A - centers and to explain them by phonon drag of electrons. Pius d and e we connect with TO phonons drag of holes in Si.

Represented in Fig. 3 the TEP temperature dependencies contain some narrow pius. The temperatures of most significant TEP pius for InSb are submitted in Table I together with Deby temperatures, energies and types of determined optically suitable phonons in InSb single crystal [12, 14]. Taking into account the data from Table I the TEP peaks in InSb may be connected with phonons participation. Identical data concerning InAs sample with data about suitable phonons in InAs single crystal [15] are submitted in Table II. Contained in the Table II data allow to connect the TEP pius in InAs with participation of phonons. It is visible from Table II that the most intensive PDE pius at temperatures 173 K and 178 K have not suitable phonons. These pius differ from the PDE pius at temperatures 208 K and 212 K on $\simeq 34K$ and may be explained by participation in the PDE effect of acoustical phonons with $T_D \simeq 34\ K$ $(2.9meV)$. It may be possible because such acoustical phonons

TABLE I: Temperatures of the TEP pius, Deby temperatures (T_D), energies (E_p) and types (Type) of suitable phonons in InSb single crystal.

T (K)	134	168	182	191	273	293	430
T_D (K)	124	170	170	170	266	280	446
E_p (meV)	5.3	14.6	14.6	14.6	22.9	24.4	16.2
Type	2TA	LA	LA	LA	LO(Γ)	LO(Γ)	2TO

TABLE II: Temperatures of the TEP pius, Debye temperatures (T_D), energies (E_p) and types (Type) of suitable phonons in InAs single crystal.

T (K)	159	173	178	196	208	212	310	320	424
T_D (K)	102	-	-	206	206	206	319	319	411
E_p (meV)	8.8	-	-	17.7	17.7	17.7	27.5	27.5	17.7
Type	TA	-	-	LA	LA	LA	TO_1	TO_1	2LA

can influence on EVC inherent oscillations, make them coherent, enable to arise coherent areas, heat superconductivity and hyperconductivity in different semiconductors [16].

One can see from Fig. 4 that temperature dependence of the TEP for InSb/GaAs contain some pius at different temperatures though according to the TEP theory similar curves for InSb and GaAs can not contain of such narrow pius. Temperature of these pius are inserted in Table III. Debye temperatures of suitable phonons energies and types of the phonons for single crystals InSb [12, 14] and GaAs [17] are inserted in the Table III also. It is visible from Table III that the temperatures of some pius TEP cannot confidently be identified with phonons in InSb and their greater number is possible to explain only by combination of such phonons. Examination of contained in Table III data enable with the more probability to refer the TEP pius in InSb/GaAs to participation of GaAs substrate phonons. The temperatures of TEP pius visible in Fig. 5 for InAs/GaAs are inserted in Table IV together with Debye temperatures and energies of suitable phonons in single crystals InAs [15] and GaAs [17]. Examination of inserted in Table IV data enable with high probability to connect

TABLE III: Temperatures of the TEP pius in InSb epitaxial layer on semiinsulator GaAs substrate (EL), Debye temperatures (T_D) , energies (E_p) and types (Type) of suitable phonons in InSb and GaAs.

EL	InSb			GaAs		
T (K)	T_D (K)	E_p (meV)	Type	T_D (K)	E_p (meV)	Type
112	-	-	-	104	9.0	TA
181	-	-	-	182	15.7	TA
253	257	22.2	TO	258	22.2	LA
315	319	-	TO + TA	336	29.0	LO
383	393	-	LO + LA	393	33.8	TO(Γ)
440	446	36.7	2LO	425	36.7	LO(Γ)
486	480	-	TO + LO	478	-	TO + LO
523	532	23.3	2TO(Γ)	540	23.3	2LA

the TEP pius in InAs/GaAs with definite role of GaAs substrate phonons.

Represented in Fig. 6 temperature dependencies of the TEP for carbon nanotube film and for silicon film on quatz substrate are similar with each other in common features. This similarity of curves A and B in Fig. 6 give definite basis to connect the pius of the curves with participation of quartz substrate phonons in phonon drag phenomenon.

D. Conclusion

Earlier the PDE in semiconductors was observed as narrow pius of the TEP at temperatures below 70 K. Taking into account the new data about the PDE pius located close to Debye temperatures of phonons the well known PDE effect at low temperatures in different materials may be explained as PDE pius caused by participation of acoustical phonons with low Debye temperatures ($T_D < 70\ K$). Absence of the PDE at T > 70 K earlier was explained by insufficiently strong coupling between electrons and phonons. Nevertheless the temperature dependence of thermoelectric power in ropes of carbon nanotubes suppos-

TABLE IV: Temperatures of the TEP pius in InAs epitaxial layer (EL) on semiinsulator GaAs substrate, Debye temperatures (T_D), energies (E_p) and types (Type) of suitable phonons in InAs and GaAs.

EL	InAs			GaAs		
T (K)	T_D (K)	E_p (meV)	Type	T_D (K)	E_p (meV)	Type
104	102	8.8	TA	104	9.0	TA
183	-	-	-	182	15.7	TA
248	-	-	-	248	21.4	LA
258	-	-	-	258	22.2	LA
267	282	24.3	LO	270	23.3	LA
326	319	27.5	TO_1	336	29.0	LO
362	383	-	LO + TA	367	31.6	TO
422	410	17.7	2LA	425	36.7	LO(Γ)

edly was explained by the PDE contribution at temperatures from 4.2 K to 300 K [18]. The narrow thermoelectric power pius observable in semiconductor materials and structures undoubtedly are connected with the phonon drag of electrons (or holes). It is necessary to take into account that the phonons frequencies can be changed or splitted under interactions between EVC and appropriate Debye temperatures can be changed. Therefore there is no basis to wait of strict concurrence of the TEP pius temperatures with Debye temperatures for phonons. The TEP pius in semiconductors single crystals are located near to Debye temperatures of phonons but in epitaxial semiconductor layers on semiinsulator substrates such pius are located near to Debye temperatures of substrate phonons. The electron-phonon coupling which supplied by electron-vibrational centers in semiconductor crystals, in epitaxial layers and else in carbon nanotube films on substrates probably is sufficiently strong for the PDE effect realization even at high Debye temperatures.

The represented experimental data specify participation in PDE effect not only long-wave acoustic phonons which are taken into account in known PDE theory but also short-wave acoustic and optical phonons in points of Brillouin zone with high density of phonons

frequencies. In connection with these data it is desirable to expand the PDE theory on a case of strong electron-phonon interaction provided by electron-vibrational centers at high temperatures.

The investigation of the PDE pius enable to study the phonons types, their energies, Debye temperatures in containing electron-vibrational centers semiconductor materials and structures.

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